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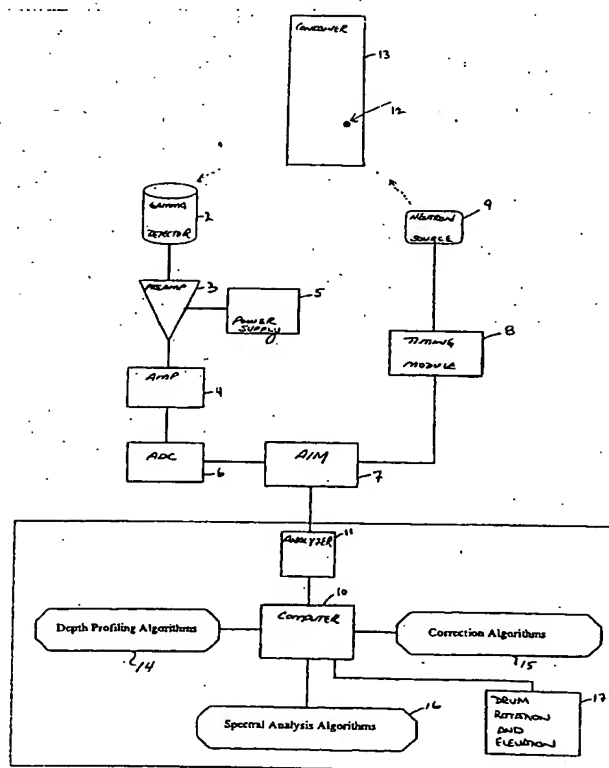
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- (54) Title: PULSED GAMMA NEUTRON ACTIVATION ANALYSIS (PGNAA) METHOD AND APPARATUS FOR NONDESTRUCTIVE ASSAY OF CONTAINERIZED CONTAMINANTS



- (57) Abstract: A PGNA method provides the nondestructive assay of a homogeneous or a non-homogeneous contaminant (50), such as a hazardous RCRA metal, in a container (36), such as a sealed 55 gallon waste drum (36). The method includes employing an interrogation chamber (40) having a wall (42) made of a neutron-reflecting material; and disposing a platform (30) within the interrogation chamber (40). At least one of rotating (31) the platform (30), and elevating (32) the platform (30) is employed. The method also includes disposing the container (36) on the platform (30) and within the interrogation chamber (40); and employing at least one neutron source (46) between the wall (42) of the interrogation chamber (40) and the container (36), with thermal neutrons (52) reacting with a container (36) and thereby effecting an emission of thermal-neutron induced gamma radiation (54) from the container (36). The container (36) is irradiated with a fast-neutron burst (44) from the neutron source (46) or as reflected by the wall (42). The fast-neutron burst (44) is characterized by a duty cycle and is thermalized and/or moderated by the container (36). Gamma radiation data is acquired (10, 11) indicative of the number or intensity of gamma rays and energies of the gamma rays during a predetermined time interval. The acquiring step includes a separate identification of thermal neutron-induced gamma rays, and the gamma radiation data is acquired over only a fraction of the duty cycle. The gamma radiation is analyzed (14, 15, 16) to detect the presence and determine the amount of the non-homogeneous contaminant (50) in the container (36).



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**PULSED GAMMA NEUTRON ACTIVATION ANALYSIS (PGNAA)
METHOD AND APPARATUS FOR NONDESTRUCTIVE ASSAY OF
CONTAINERIZED CONTAMINANTS**

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BACKGROUND OF THE INVENTION

Field of the Invention

This invention pertains generally to PGNAA (*i.e.*, pulsed gamma neutron activation analysis) and, more particularly, to methods and apparatus which apply PGNAA to the nondestructive assay of contaminants in containers.

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Background Information

U.S. Patent No. 5,539,788 discloses a Pulsed Gamma Neutron Activation Analysis (PGNAA) system for determining depth profiles of concentrations of hazardous elements in soils. The system includes a neutron source for generating neutrons of a first energy level and irradiating a volume of soil with the neutrons.

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Nuclear reactions are effected within the soil and gamma radiation is emitted from the soil. The system also includes an array of gamma detectors for detecting gamma radiation emitted from the soil; source electronics for controlling the width of regularly repeated pulses of neutrons generated by the neutron source; detector electronics associated with the gamma detectors for amplifying and digitizing signals generated by the gamma detectors and storing data representing the digitized signals; spectral analysis software for analyzing the data and determining the concentrations of selected target elements in the soil; and an acquisition interface module (AIM). The AIM controls the timing of the source and detector electronics, such that the neutron source generates neutrons in regularly repeated bursts of a prescribed pulse width, and the detectors and detector electronics detect gamma rays during a plurality of time intervals associated with the burst and acquire groups of data. Each group is indicative of a number of gamma rays emitted and their energy levels during a corresponding time interval. The

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system also includes a platform carrying the neutron source, an array of gamma ray detectors, and software for determining a depth profile of a target element.

U.S. Patent No. 5,781,602 discloses PGNAA to determine *in situ* the content of nickel, copper, or other embrittlement-enhancing elements within the weld metal joints of nuclear reactor pressure vessels. Patent 5,781,602 states that, as disclosed in Patent 5,539,788, PGNAA provides the capability for *in situ* measurement of concentrations of contaminant elements in materials at levels as low as tens of parts per million for elements such as mercury or cadmium, within minutes to hours.

U.S. Patent No. 5,825,030 discloses a bulk material analyzer employing PGNAA in which bulk material is received in an activation region between a pair of neutron sources laterally disposed on one side of the activation region for emitting neutrons for bombarding the bulk material within the activation region to cause gamma-rays to be emitted from the bombarded bulk material and a pair of gamma-ray detectors laterally disposed on an opposite side of the activation region for detecting gamma-rays emitted from the bulk material. A neutron moderator is disposed between the neutron sources and the activation region for reducing the velocity of the emitted neutrons such that the neutrons entering the activation region include thermal neutrons and faster neutrons. The uniformity of sensitivity of the bulk material analyzer to materials at different positions in a sample of the bulk material being analyzed is enhanced by controlling the proportions of thermal neutrons at different positions within the sample being analyzed. A neutron absorber disposed close to the one side of the activation region reduces the proportion of thermal neutrons that enter the activation region from the one side of the activation region. A neutron reflector disposed on the opposite side of the activation region, at least as close to the activation region as the gamma-ray detectors, reflects neutrons into the activation region for bombarding bulk material within the activation region. In some embodiments, the neutron absorber and/or the neutron reflector are adjustable; and the disposition of the neutron sources also is adjustable for adjusting the proportion of the thermal neutrons that enter the activation region from the one side of the activation region.

It is known to employ PGNAA for the nondestructive assay (NDA) of certain types of contaminants (*e.g.*, metals defined as hazardous under the Resource Conservation and Recovery Act (RCRA)) (40 CFR §§ 260-70) in sealed containers. Dulloo, A. R., et al., "Detection Limits of a Laboratory Pulsed Gamma Neutron

Activation Analysis System for the Nondestructive Assay of Mercury, Cadmium, and Lead," Nuclear Technology, vol. 123, July 1998, pp. 103-12, discloses the ability of a laboratory PGNA system to detect and quantify a uniform distribution (*i.e.*, a homogeneous distribution in a concrete matrix) of RCRA metals (*e.g.*, mercury, cadmium and lead) in surrogate waste matrices contained in eight-gallon drums.

It is known to employ a portable isotopic neutron spectroscopy (PINS) chemical assay system as a non-destructive evaluation tool to identify the contents of munitions and chemical storage containers. See Caffrey, A. J., et al., "U.S. Army Experience with the PINS Chemical Assay System", EGG-NRP-11443, September 1994, pp. 1-24.

Neutron-induced reactions can be divided into two broad categories, threshold reactions and exoergic reactions. For threshold reactions, energy in the form of neutron kinetic energy is required to supply a certain minimum energy to the reaction system before the reaction can proceed. Neutrons with energies below this minimum threshold energy are incapable of inducing the nuclear reaction. For exoergic reactions, the threshold is zero; that is, neutrons with all energies are capable of inducing the reaction. Since neutrons lose energy via nuclear collisions, the minimum energy possible for a neutron is determined by the thermal motion of the atoms in the stopping medium. Neutrons with this minimum average energy are referred to as thermal neutrons and have a mean energy of approximately 0.0252 eV at room temperature.

In the process of neutron activation, a neutron of energy E collides with the nucleus of an atom in the sample and initiates a reaction. For a neutron of thermal energy, the reaction might be absorption of the neutron into the nucleus, creating the next higher mass isotope of that element. If the neutron is more energetic (*e.g.*, with several million electron volts (MeV) of kinetic energy), other nuclear reactions are possible. These other reactions include inelastic scattering from the nucleus, exciting the nucleus according to its internal structure of quantum energy levels, or other reactions (*e.g.*, (n,p), (n, alpha), (n,2n)) in which nuclear transmutation to another element occurs. In each of these cases, the residual nucleus is left in a highly excited internal state, and decays to its ground state almost instantaneously (10^{-14} s or less), emitting a gamma ray of several MeV of energy. The energy of this gamma ray is uniquely characteristic of the quantum structure of the residual nucleus, and thus is a signature of the original target nucleus. The number of atoms of each of the elements of

interest in a sample can be estimated by detecting and collecting the spectrum of gamma rays emitted by the sample and integrating the appropriate peaks.

The PGNAA process is governed by the following equation:

$$A = N\sigma\phi\beta$$

5 wherein:

A = disintegrations per second producing the desired gamma rays,

N = the number of target nuclei for the reaction,

σ = the reaction cross section (10^{-24} cm²),

ϕ = the flux of neutrons of the required energy (cm⁻²sec⁻¹), and

10 β = the branching ratio (*i.e.*, a fraction between 0 and 1 indicating how often this capture produces the gamma ray of interest).

There is room for improvement in nondestructive assay technologies, primarily because of a need to detect lower concentrations of contaminants than presently possible with existing technologies.

15 SUMMARY OF THE INVENTION

While the assay of uniformly contaminated waste is relatively straightforward, the assay of waste having a nonuniform distribution of contaminant can be difficult, especially when the waste is located in a large container. The present invention provides improvements in PGNAA methods and apparatus for nondestructive
20 assay of a non-homogeneous contaminant in a container. The invention also provides improvements in a PGNAA method for nondestructive assay of a non-homogeneous contaminant or a homogeneous contaminant in a container.

In accordance with the invention, a PGNAA method for nondestructive assay of a non-homogeneous contaminant in a container comprises: (a) employing an
25 interrogation chamber having a wall made of a neutron-reflecting material; (b) disposing a platform within the interrogation chamber; (c) employing at least one of: (i) rotating the platform, and (ii) elevating the platform; (d) disposing the container on the platform and within the interrogation chamber; (e) employing at least one neutron source between
the wall of the interrogation chamber and the container; (f) irradiating the container with
30 a fast-neutron burst from the neutron source or as reflected by the wall, the fast-neutron burst characterized by a duty cycle and being thermalized and/or moderated by the container, with thermal neutrons reacting with the container and thereby effecting an emission of thermal-neutron induced gamma radiation from the container; (g) acquiring

gamma radiation data indicative of the number or intensity of gamma rays and energies of the gamma rays during a predetermined time interval, the acquiring step including a separate identification of thermal neutron-induced gamma rays, and wherein the gamma radiation data is acquired over only a fraction of the duty cycle; and (h) analyzing the
5 gamma radiation data to detect the presence and determine the amount of the non-homogeneous contaminant in the container.

Preferably, both of the steps of rotating the platform, and elevating the platform are employed. The analyzing step may include interference-correction to detect and/or correct for: (a) signal interference from gamma rays emitted by other
10 elements present in the container, and (b) perturbation of interrogating neutron flux resulting from neutron-producing fissile materials and neutron poisons in the container. The analyzing step may also include computer modeling to perform gamma spectral deconvolution, a gamma spectral analysis program to enhance system sensitivity for elemental detection, and adjoint transport calculations to infer contaminant distribution
15 through pattern recognition.

A plurality of neutron sources may be employed in order to produce significant thermal neutron fluxes at different average depths in the container, with the analyzing step including depth profiling analysis of an element detectable by thermal neutron activation.

20 As another aspect of the invention, a PGNA method for nondestructive assay of a non-homogeneous contaminant or a homogeneous contaminant in a container, comprises: (a) employing an interrogation chamber having an annular wall made of a neutron-reflecting material, the wall having a first portion and a second portion; (b) disposing a neutron-moderating material on the first portion of the wall of the
25 interrogation chamber; (c) disposing a platform within the interrogation chamber; (d) employing at least one of: (i) rotating the platform, and (ii) elevating the platform; (e) disposing the container on the platform and within the interrogation chamber; (f) employing a plurality of neutron sources between the wall of the interrogation chamber and the container; (g) irradiating the container with a fast-neutron burst from at least one
30 of the neutron sources or as reflected by the second portion of the wall, the fast-neutron burst characterized by a duty cycle and being thermalized and/or moderated by the container, with thermal neutrons reacting with the container and thereby effecting an emission of thermal-neutron induced gamma radiation from the container; (h) irradiating

the container with a thermal-neutron burst from at least one of the neutron sources as moderated by the neutron-moderating material on the first portion of the wall of the interrogation chamber and as reflected by the first portion of the wall, the thermal-neutron burst characterized by a duty cycle, with thermal neutrons reacting with a first layer of the container and thereby effecting an emission of thermal-neutron induced gamma radiation from the first layer; (i) selectively acquiring: (1) thermal-neutron induced gamma radiation from the container, or (2) thermal-neutron induced gamma radiation from the first layer, to provide gamma radiation data indicative of the number or intensity of gamma rays and energies of the gamma rays during a predetermined time interval, the selectively acquiring step including a separate identification of thermal neutron-induced gamma rays, and wherein the gamma radiation data is acquired over only a fraction of the duty cycle; and (j) analyzing the gamma radiation data to detect the presence and determine the amount of the non-homogeneous contaminant or the homogeneous contaminant in the container.

As a further aspect of the invention, a PGNAA apparatus for nondestructive assay of a non-homogeneous contaminant in a container comprises: an interrogation chamber having a wall made of a neutron-reflecting material; a platform for supporting the container within the interrogation chamber; means for rotating and/or elevating the platform; at least one neutron source between the wall of the interrogation chamber and the container; means for irradiating the container with a fast-neutron burst from the neutron source or as reflected by the wall, the fast-neutron burst characterized by a duty cycle and being thermalized and/or moderated by the container, with thermal neutrons reacting with the container and thereby effecting an emission of thermal-neutron induced gamma radiation from the container; at least one gamma ray detector; means for acquiring gamma radiation data from the gamma ray detector indicative of the number or intensity of gamma rays and energies of the gamma rays during a predetermined time interval, including a separate identification of thermal neutron-induced gamma rays, and wherein the gamma radiation data is acquired over only a fraction of the duty cycle; means for analyzing the gamma radiation data to detect the presence and determine the amount of the non-homogeneous contaminant in the container.

The gamma detector may include a shield, such as a ^6Li -bearing shield. Preferably, the ^6Li -bearing shield is disposed around the gamma detector, and a moderating and/or shielding material is disposed around the ^6Li -bearing shield.

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BRIEF DESCRIPTION OF THE DRAWINGS

A full understanding of the invention can be gained from the following description of the preferred embodiments when read in conjunction with the accompanying drawings in which:

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Figure 1 is a block diagram of a PGNA system in accordance with the present invention;

Figure 2 is a block diagram of a PGNA drum assay chamber for a PGNA system in which the container has a homogeneous concentration of a contaminant;

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Figure 3 is a block diagram of a PGNA drum assay chamber for the PGNA system of Figure 1 in which the container has a non-homogeneous concentration of a contaminant;

Figure 4 is a block diagram of a PGNA drum assay chamber for the PGNA system of Figure 1 in which such system is employed for either homogeneous or non-homogeneous assay;

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Figure 5 is a top view of another embodiment of a PGNA drum assay chamber for the PGNA system of Figure 1 in which such system is employed for either homogeneous or non-homogeneous assay;

Figure 6 is an elevational view of the interrogation chamber and gamma detector assembly of another PGNA system in accordance with the present invention;

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and

Figure 7 is a cross-sectional view along lines 7-7 of Figure 6.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

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Improved nondestructive technologies that are capable of detecting hazardous materials are needed for the assay of waste drums and other containers. For example, such technologies may be used to screen out waste containers destined for waste treatment processes that contain high concentrations of elements that are known either to result in effluent stream concentrations which exceed regulatory limits, or to chemically interfere with the control of a particular treatment process. The technologies

may also be used to screen containers destined for repositories such as the Waste Isolation Pilot Plant (WIPP), which is an underground repository licensed to safely and permanently dispose of transuranic radioactive waste left from the research and production of nuclear weapons.

5 Referring to Figure 1, a PGNA system 1 includes a suitable gamma detector 2 (*e.g.*, a high-performance germanium detector, cadmium zinc telluride detector, bismuth germanate detector, sodium iodide detector, lead bromide detector, lead iodide detector, or other suitable detector); a suitable high speed, high throughput count rate preamplifier (PREAMP) 3; a high count rate, high speed spectroscopy
10 amplifier (AMP) 4; a detector high voltage power supply 5; a high speed analog-to-digital converter (ADC) 6; an Acquisition Interface Module (AIM) 7; a special neutron generator timing module 8; and a pulsed neutron source or generator 9 (*e.g.*, pulsed D-T (deuterium-tritium) or D-D (deuterium-deuterium) accelerator). The timing module 8 provides for time-correlated data acquisition in order to coordinate firing of the neutron
15 generator 9 with gamma pulse acquisition. The system 1 also includes suitable computer 10, such as a high-speed mini-computer, and a multi-channel analyzer software 11, which is typically integrated with or loaded on the computer 10, for data acquisition of gamma radiation data from the gamma ray detector 2, and analysis and system control. An example of suitable system components 2-11 for the system 1 is
20 disclosed in Patent 5,539,788, which is incorporated by reference herein.

A suitable time-correlated data acquisition technique is disclosed in Dulloo, A. R., et al., Nuclear Technology; and Patent 5,539,788. For example, a time-correlated PGNA data acquisition technique allows the temporal separation of: (a) gamma rays from fast-neutron induced events from (b) gamma rays from thermal-
25 neutron induced events and from (c) gamma rays from decaying radioactive species, thereby allowing analysis of each type of gamma rays with superior signal/noise ratio. A suitable delay time for this technique is determined by empirical data for the chamber (not shown) for the container 13, in order to provide the optimum signal-to-noise ratio.

It will be appreciated that while reference has been made to the
30 exemplary computer 10, a wide range of other processors such as, for example, mainframe computers, workstations, personal computers (*e.g.*, network or standalone), microcomputers, and other microprocessor-based computers may be employed.

The exemplary PGNAA-based system 1 is employed in accordance with the present invention to provide the nondestructive assay (NDA) of a non-homogeneous contaminant 12 in a container 13. For example, the contaminant 12 may be a hazardous metal, in the container 13, such as a sealed container, a 55-gallon waste drum, or other types of sealed waste containers for waste matrices that are not limited to homogeneous concentrations of contaminants. The system 1 employs innovative adaptations of conventional PGNAA and has features that allow elemental detection of both homogeneous and non-homogeneous distributions of element(s) of interest.

Applications of the system 1 include, but are not limited to, one or more RCRA metals (e.g., mercury, cadmium, lead) and/or other hazardous elements analyzed in mixed-waste drums and chlorine content analysis of sealed containers destined for treatment processes. Other applications include noninvasive analysis of hydrogen and/or boron content of nuclear fuel, and noninvasive analysis of drums containing scrap metal (e.g., stainless steel) destined for recycling processes in order to verify that the content of the drum is consistent with the content as indicated on the drum label.

Gamma spectra are collected and stored on the computer 10. Suitable post-processing computer algorithms 14,15,16 are employed to analyze the spectral data in conjunction with the analyzer 11. These algorithms include, in accordance with the present invention, depth profiling algorithms 14 for suitable depth profiling analysis; correction algorithms 15 for suitable interference-correction, and spectral analysis algorithms 16 for both fast-neutron induced decay gamma analysis and passive gamma analysis. The computer 10 also includes suitable rotation and elevation control algorithms 17 (as discussed below in connection with Figures 2-6) for rotating and elevating the container 13.

Although the exemplary system 1 is disclosed in terms of the assay of metals defined as hazardous by RCRA in an exemplary 55-gallon drum, this system 1 may be applied to the assay of any trace element that has a suitable cross section for a neutron interaction that leads to the emission of at least one gamma ray with sufficiently high yield. The system 1 may also be applied to a wide range of different containers.

The exemplary depth profiling algorithms 14 are adapted from the techniques disclosed in Patent 5,539,788 (for soil analysis). Two methods for determining the depth distribution of contaminants are disclosed in Patent 5,539,788: (1) employing the known behavior of the neutron energy distribution with depth as the

neutrons propagate beneath the surface; and (2) employing the known attenuation properties of a substance for gamma rays of very different energies, emitted with known yields, from the same substance.

In the exemplary embodiment, the use of a depth profiling technique is employed to infer contaminant distribution in relatively large containers (e.g., 55-gallon drums). Preferably, to achieve a depth profile of an element detectable by thermal neutron activation, a plurality (e.g., two to four sources may typically be employed) of the exemplary neutron sources 9 are employed, which produce significant thermal neutron fluxes at very different average depths in the container 13. For the case of a plurality of neutron sources 9, the neutron sources need not be identical. For example, one source can be a D-T source, which emits 14-MeV neutrons, while the other can be a D-D source, which emits 2.47-MeV neutrons.

The method to determine whether a contaminant is present within a prescribed depth range is disclosed in Patent 5,539,788, and is achieved in one (or more than one) of three ways.

One way is to employ different neutron sources, and threshold nuclear reactions. Some contaminant elements (e.g., Pb) can be identified through γ -rays generated as a result of a threshold reaction. As the neutrons travel through the matrix material, they lose kinetic energy as a result of successive scattering collisions. Therefore, for a given energy of neutrons emitted by the source 9, there is a maximum depth in the sample, beyond which the neutrons from the source would no longer retain sufficient kinetic energy to initiate the threshold nuclear reaction in the contaminant element. This "maximum depth" is readily calculated using neutron transport codes. Therefore, if that source is used to irradiate the sample, and signature γ -rays from the threshold reaction of the contaminant element are produced, the contaminant concentration so detected must lie at a depth not greater than this maximum depth. There may still be contaminant located at greater depths, but those contaminant atoms will not contribute to the system response, which will measure the concentration present only as far down as the maximum depth for inducing the threshold reaction.

For example, in the case of the detection of lead through the (n,n') reaction on ^{207}Pb , the 1.063 MeV γ -ray can be generated from neutrons of kinetic energy greater than or equal to approximately 1.1 MeV. This energy can be regarded as a threshold energy to generate 1.063 MeV γ -rays. This threshold energy is clearly

available if either a D-D neutron source (neutron energy = 2.47 MeV) or a D-T neutron source (neutron energy = 14 MeV) is employed. However, for the D-T source, the maximum depth at which the neutrons could initiate the reaction producing the 1.063 MeV γ -ray is greater than the maximum depth at which neutrons from the D-D source could initiate the reaction. Each source would, thus, potentially be able to supply information on the concentration of lead down to a different maximum depth, and a coarse depth distribution could be inferred.

A second way is to employ different neutron sources, and thermal neutron capture of nuclear reactions. As discussed above, the loss of neutron kinetic energy as a result of successive collisions leads to a typical depth at which neutrons starting with a particular initial kinetic energy will reach some lower kinetic energy of interest. Thus, a pulse of 14 MeV neutrons will achieve thermal energy (approximately 0.025 eV) at a depth into the sample which is deeper than that at which a source neutron of lesser kinetic energy (e.g., 2.47 MeV) will achieve thermal energy. Hence if a D-T source and then a D-D source are consecutively employed to determine the concentration of a particular element (e.g., Cd or Hg) using a thermal neutron capture reaction, the depth into the sample being interrogated by the flux of thermal neutrons will be different for the two sources. Once again, the average concentration is extracted for two different depths into the sample, and a coarse depth distribution of the contaminant can be obtained.

A third way is to employ differential attenuation of signature γ -rays. Patent 5,539,788 discloses how to extract a coarse depth distribution of a contaminant using a nuclear reaction, provided the reaction causes the contaminant to emit a significant quantity of γ -rays of different energies. Briefly, the intensity of γ -rays of different energies undergoes differing degrees of attenuation by the matrix material as they travel back toward the detector, to re-emerge from the sample. Thus, the detected ratio of the intensities of these two (or more) γ -rays will vary with the depth in the sample at which the emitting atom was located. By grouping these ratios into bins, it is possible to mathematically invert this information in order to infer how much contaminant was in each of the (two or more) regions of depth in the sample.

This method, as taught in Patent 5,539,788, utilizes calibration measurements to establish the γ -ray responses expected from contaminants at different depths. An alternate approach to the deconvolution of a contaminant depth profile from

the measured data involves the generation of adjoint functions, using typical neutron- and γ -ray transport codes. These functions provide the detector response as a function of the concentration of contaminant at various locations. By trial and error, these adjoint functions can be used with postulated contaminant distributions, until a
5 distribution which matches the measured γ -ray intensities is identified.

As discussed above, Patent 5,539,788 discloses a method to compute a depth at which the neutrons have energy appropriate to initiate a reaction that produces gamma radiation indicative of the contaminant, and to generate depth profile data representing the amount of the contaminant at a plurality of depth ranges in the
10 container.

The exemplary correction algorithms 15 provide interference-correction to detect and/or correct for: (1) signal interference from gamma rays emitted by other elements (*i.e.*, other than the non-homogeneous contaminant 12) present in the container 13; and (2) perturbation of the interrogating neutron flux from the neutron source 9
15 resulting from neutron-producing fissile materials and neutron poisons (*e.g.*, boron and chlorine in the waste matrix) in the container 13. An example of a suitable method for the correction of gamma interference is disclosed in Dulloo, A. R., et al., Nuclear Technology. A specific example of a method to detect and correct for neutron perturbation (item 2, above) as presented, for example, by boron includes: (a)
20 monitoring the presence and intensity of the signature gamma ray emitted by boron as a result of thermal neutron capture in the PGNA spectrum; (b) if the intensity of the boron gamma ray is above an empirically predetermined threshold, then perturbation of the neutron flux is indicated; and (c) a suitable algorithm is applied to the gamma spectral data to correct for the neutron flux perturbation. This algorithm relates boron
25 gamma-ray intensity to neutron flux attenuation using factors derived from a priori measurements (assisted by modeling calculations) made with known boron concentrations in a matrix similar to that of the sample matrix. This correction method is matrix-specific, and is only effective for boron concentrations that do not present significant self-shielding effects. A similar method to detect and correct for neutron
30 perturbation as presented, for example, by chlorine may be employed.

The exemplary spectral analysis algorithms 16 preferably include computer modeling to perform gamma spectral deconvolution and, in particular, a gamma spectral analysis program to enhance system sensitivity for elemental detection.

Preferably, the system 1 employs a plurality of the sources 9 and a plurality of the detectors 2. This improves measurement statistics and, in the case of nonuniform distributions, obtains spatial information, which is well known for passive gamma analysis, on contaminant distribution through the separate analysis of signals from individual detectors that are located so as to observe overlapping spatial regions. An example of a gamma spectral analysis program that is commercially available is Canberra Industries' Genie-2000 Gamma Analysis Software.

Furthermore, the exemplary spectral analysis algorithms 16 preferably employ adjoint transport calculations, to infer contaminant distribution through pattern recognition. Bell, G.I., et al., "Nuclear Reactor Theory," Van Nostrand Reinhold Company, 1970, pp. 252-314, discloses general adjoint calculation methodology. As a specific example, for the case of two detectors 2 situated at different locations in an interrogation chamber (not shown) for the container 13, there are overlapping viewing regions. Due to the different attenuation of the signature gamma rays reaching the detectors 2, positional information on the contaminant 12 may be obtained through adjoint functions. These functions provide the detector response as a function of the concentration of contaminant at various locations. By trial and error, these adjoint functions may be used with postulated contaminant distributions, until a distribution which uniquely matches the measured γ -ray intensities for both detectors 2 is identified.

Figure 2 shows a block diagram of the exemplary system 1 of Figure 1 as applied to a waste drum 18 having a homogeneous distribution of a contaminant. An important component is the layout and composition of the wall of the interrogation chamber 20. The neutron source 9 is typically located within a cavity defined by the chamber walls, whereas the detector 2 is typically located within a penetration through the chamber wall adjacent to the source 9. If the system 1 is to be configured solely for the assay of uniformly (*i.e.*, homogeneously) contaminated samples, then the entire chamber annular wall is preferably made of a neutron-moderating material 22 with strong neutron-moderating properties. Examples of such materials include polyethylene, water (*i.e.*, H_2O or D_2O), and graphite. If using polyethylene, a high-density type is preferred. The thickness of the polyethylene wall is typically between about 5 cm and 20 cm. It is also advantageous to place a layer 24 of a highly neutron-reflective material, such as, for example, graphite or bismuth, around the exterior of the wall of the chamber 20. High-purity grade (*e.g.*, reactor grade) graphite is normally

employed for this application. The thickness of a graphite reflector 24 is typically between about 10 cm and 100 cm, although a greater thickness may be employed.

By placing one or more detectors 2 and one or more neutron sources 9 on the same side of the sample waste drum 18, the gamma rays contributing primarily to the detector response will originate from a surface layer 26 of the sample 28 in such drum. Since the sample 28 is homogeneously contaminated, the surface layer 26 is representative of the entire sample.

A suitable platform 30 is disposed within the interrogation chamber 20 and is employed to provide support, rotation and elevation of the drum 18 within the chamber 20 in order to analyze multiple segments of the sample 28. This is desired in order to confirm that the sample 28 is indeed homogeneously contaminated.

The rotation and elevation control algorithms 17 of Figure 1 are employed to control the platform 30 of Figure 2, which includes both a rotating turntable 31 as well as an elevating platform portion 32 in order to rotate/elevate the drum 18 during interrogation to assay different sections thereof.

Rotational speed typically ranges from 0 to 10 revolutions per minute (rpm), whereas the elevation speed is dictated by the assay time required for sample measurement. Typically, for a 55-gallon drum that is assayed for 600 s, the elevation speed ranges from about 0 to 10 cm per minute. The elevation motion of the drum 18 can be either stepped (*e.g.*, a 360 degree rotation followed by a step change in elevation) or continuous (*e.g.*, a helical path of both rotation and elevation which concurrently rotates and elevates the platform), depending on the type of elevating mechanism.

To address a sample distribution that is radially homogeneous (*i.e.*, along a radius 33) but axially nonuniform (*i.e.*, along rotation axis 34), then the same configuration of the chamber 20 for homogeneous samples may be employed. The drum 18 can be elevated vertically by the platform 30 in order that axial segments of the drum content can be analyzed sequentially, and an axial profile of the contaminant distribution is generated.

Alternatively, sequential assays taken with the detector 2 and the source 9 placed at different positions along the length of the drum 18 may be done to generate the axial contaminant profile in such drum. Collimation of the detector 2 may be employed to improve spatial resolution in both cases.

Referring to Figure 3, for drums, such as 36, that contain an arbitrary, non-homogeneous distribution of a sample 38, a chamber 40 having a wall 42 made of a material that has good neutron reflective properties, but not particularly strong moderating properties, is employed. Energetic source neutrons 44 from one or more fast neutron sources 46 are then reflected by neutron scattering reactions 48 through air space 49 and into the sample 38 with relatively higher kinetic energies and are, therefore, able to penetrate deeper into such sample before being thermalized (*i.e.*, their energies are reduced to thermal energy). Hence, there is a greater likelihood that a non-homogeneous contaminant, such as 50, located deep into the sample 38, may be interrogated by thermal neutrons 52 and produce thermal neutron-induced signature gamma rays 54 that are then detected by the one or more gamma detectors (GD) 56. This method assumes that the matrix inside the drum 36 is capable of neutron moderation. For drums that contain matrices that are substantially non-moderating (*e.g.*, a matrix filled with air voids), the method employed to assay homogeneous contaminants can be applied as the transport of thermal neutrons to deep locations within these types of matrices is relatively unimpeded provided that the matrix is also non-absorbing.

In summary, Figure 3 shows the interrogation of the drum 36 with a non-homogeneous contaminant distribution of the sample 38 using a non-moderating or suitably lightly-moderating neutron reflecting wall material 42. The reflected source neutrons 44 are still energetic and are able to travel to considerable depths into the drum 36 before being thermalized. Neutrons thermalized in the vicinity of the contaminant 50 (*e.g.*, at 58) interact with the contaminant 50 to produce signature gamma rays 54 that can then be detected by the gamma detectors 56. The drum 36 is irradiated with a fast-neutron burst from the neutron source(s) 46 or as reflected by the wall 42. Such fast-neutron burst is characterized by a duty cycle and is thermalized and/or moderated by the drum 36 and the sample 38. The thermal neutrons 52 react with the sample 38 and the contaminant 50 of the drum 36 and thereby effect an emission of thermal-neutron induced gamma radiation 54 from the drum 36. The system 1 of Figure 1 acquires gamma radiation data indicative of the number or intensity of the gamma rays 54 and energies of such gamma rays 54 during a predetermined assay time interval. The gamma radiation data are acquired over only a fraction of the duty cycle of the fast-

neutron burst. This acquisition includes a separate identification of the thermal neutron-induced gamma rays.

For example, the platform 30 may be raised to elevate the drum 18 of Figure 2, in order to assay different vertical sections S1,S2,S3 thereof. Also, the platform 30 may be rotated to rotate the drum 36 of Figure 3, in order to assay different radial sections R0,R1,R2,R3 thereof. Further analysis of the drums 18,36 may be performed by both rotating and elevating the platform 30.

In accordance with the present invention, the gamma radiation data are analyzed by the algorithms 14,15,16 of Figure 1 to detect the presence and determine the amount of the non-homogeneous contaminant 50 in the drum 36. The axial segment of the drum 36 where the contaminant is located is easily determined by correlating the time elapsed from the start of the assay when the signature gamma ray is detected with the drum elevation speed. Empirically-determined calibration constants coupled with adjoint calculations are then used to relate gamma intensity to the contaminant concentration in that segment.

The use of passive gamma analysis for the assay of naturally radioactive contaminants (e.g., ^{238}U , ^{235}U , ^{232}Th , ^{137}Cs) through the detection and analysis of passive gamma rays emitted by the decay of these contaminants to provide the concentrations of these radioactive nuclides.

A suitable technique for fast-neutron induced decay gamma analysis is disclosed in Dulloo, A. R., et al., Nuclear Technology. For example, the assay of certain types of contaminants, such as lead, is possible through detection of decaying gamma rays from short-lived nuclides produced by fast-neutron induced reactions.

If multiple signature gamma rays 54 from the same contaminant 50 of Figure 3 are emitted, then the depth of such contaminant in the sample 38 can be inferred by the depth profiling algorithms 14 of Figure 1 by employing the depth profiling technique as disclosed in Patent 5,539,788. Furthermore, spatial information is also obtained by the spectral analysis algorithms 16 of Figure 1 from the analysis of spectral data acquired with plural gamma detectors 56 placed at different locations around the drum 36.

While two separate chambers 20,40 (i.e., one for homogeneous samples 28 of Figure 2, and the other for non-homogeneous samples 38 of Figure 3) can be employed, if desired, cost issues may favor the single chamber 60 as shown in Figure 4.

The chamber 60 is suitable for both types of distributions. One half of the wall 62 of the chamber 60 (*i.e.*, toward the left side of Figure 4) is made of both a moderator (M) material 64 and a reflector (R) material 66, in order to optimize for homogeneous-distribution assaying in that part of the chamber 60. The other half of the chamber wall 62 (*i.e.*, toward the right side of Figure 4) is made of only the reflector (R) material 66, in order to optimize for non-homogeneous-distribution assaying.

Through rotation/elevation of the waste drum 68 (as discussed above in connection with Figure 3), the strategic placement of neutron sources 70 and gamma detectors 72 at different locations around the drum 68 in the chamber 60, and the analytical techniques discussed above in connection with Figures 1-3, analysis of both types of distributions are possible with the system 1 and the single chamber 60. Thus, the selective use and strategic placement of the neutron-moderating (M) and/or neutron-reflective (R) materials 64,66 in the configuration of the assay system 1 enhances both the interrogation of the sample contents of the drum 68 located close to the drum walls (*i.e.*, by thermal-neutrons) in one part of the chamber 60, and, in a different part of such chamber, interrogation of contents 74 located at greater depths into the drum 68 (*i.e.*, by increasing the fast-neutron component of the neutron flux incident on such drum).

In summary, the drum 68 is irradiated with a fast-neutron burst from one or both of the neutron sources 70 or as reflected by the second portion (*i.e.*, the right portion of Figure 4) of the wall 62. The fast-neutron burst is characterized by a duty cycle and is thermalized and/or moderated by the drum 68, with thermal neutrons reacting with the contents 74 of the drum 68 and thereby effecting an emission of thermal-neutron induced gamma radiation therefrom. The drum is also irradiated with a thermal-neutron burst from one or both of the neutron sources 70 as moderated by the neutron-moderating material (M) 64 on the first portion (*i.e.*, the left portion of Figure 4) of the wall 62 of the interrogation chamber 60 and as reflected by that first portion of the wall. The thermal-neutron burst is characterized by a duty cycle, with thermal neutrons reacting with a first layer 76 of the drum 68 and thereby effecting an emission of thermal-neutron induced gamma radiation therefrom. In turn, the system 1 of Figure 1 is employed to selectively acquire thermal-neutron induced gamma radiation from the contents 74 of the drum 68, or thermal-neutron induced gamma radiation from the first layer 76 to provide gamma radiation data indicative of the number or intensity of gamma rays and energies of the gamma rays during a predetermined time interval.

The system 1, in combination with the chamber 60 of Figure 4, also analyzes the gamma radiation data to detect the presence and determine the amount of the non-homogeneous contaminant 74 in the drum 68. Also, the system 1, in combination with the chamber 60 of Figure 4, may detect the presence and determine the amount of a homogeneous contaminant in the drum 68 (e.g., the homogeneous sample 28 of Figure 2).

Alternatively, rather than disposing no neutron-reflective material (as shown in Figure 4) on the second portion of the wall 62 opposite the first portion having the neutron-moderating (M) material 64, as shown in Figure 5, the chamber 80 includes a wall 81 made of a reflector (R1) material 82. Disposed on one half of that reflector material 82 (*i.e.*, toward the left side of Figure 5) is a moderator (M) material 84, and disposed on the other half of the reflector material (R1) 82 (*i.e.*, toward the right side of Figure 5) is a neutron-reflecting material (R2) 86 on the second portion of the wall 81 opposite the first portion having the reflector (R1) material 82.

Figures 6 and 7 show another PGNA system 90, which includes an improved gamma detector assembly 92 in accordance with the present invention. Although the chamber of the system 90 is similar to the chamber 20 of Figure 2, the improved gamma detector assembly 92 is applicable to the chambers 40, 60, and 80 of respective Figures 3, 4 and 5.

As shown in Figure 7, the detector assembly 92 includes an exemplary Ge crystal 94, which is suitably mounted in the detector housing 96. A conventional ^6Li -bearing thermal neutron shield 97 is disposed around the crystal-containing region of the housing 96. A moderating and/or shielding material 98, such as the exemplary tungsten material, is disposed around the shield 97 and substantially around the housing 96. An example of a suitable ^6Li shield material is lithium-6 fluoride (^6LiF). Examples of suitable moderating/shielding materials are bismuth and tungsten. The ^6Li material absorbs thermal neutrons that would otherwise enter the detector and produce gamma rays through activation reactions. ^6Li has the advantage that it absorbs thermal neutrons without producing reaction gamma rays. The moderating/shielding material 98 serves two purposes: (1) shielding the detector assembly 92 from gamma radiation originating from regions other than the waste drum 100; and (2) slowing down fast neutrons to diminish the neutron interactions with the detector. The degree of "slowing down" depends upon the exact constitution of the moderating/shielding material 98. Energetic

neutrons will cause radiation damage in the detector, so the moderating/shielding material 98 prolongs the lifetime of the Ge crystal 94. Furthermore, gamma rays produced by fast and thermal neutrons in the gamma ray detector are diminished by the presence of the moderating/shielding material 98. Other suitable neutron shield isotopes include ^{10}B (e.g., $^{10}\text{B}_4\text{C}$, Zr^{10}B , $\text{H}_3^{10}\text{BO}_3$) or cadmium or gadolinium in metallic or oxide form. These materials have the disadvantage that they produce gamma rays when thermal neutrons are absorbed. Other suitable moderating/shielding materials include lead, tantalum, platinum, depleted uranium, or other suitable heavy elements that do not strongly absorb thermal neutrons and/or produce high yields of neutron-induced gamma rays.

The exemplary arrangement of the detector shield 97 is to have ^6Li material around the detector housing 96, and the moderating/shielding material 98 around the ^6Li material. Another arrangement is to have an additional layer of ^6Li material around the moderating/shielding material 98.

A suitable transmission gamma ray source 102 is provided to evaluate the density composition of the waste drum contents. Suitable transmission sources include ^{137}Cs , ^{60}Co , ^{22}Na , ^{152}Eu , or other suitable high-energy gamma ray emitting isotope.

While specific embodiments of the invention have been described in detail, it will be appreciated by those skilled in the art, that various modifications and alternatives to those details could be developed in light of the overall teachings of the disclosure. Accordingly, the particular arrangements disclosed are meant to be illustrative only, and not limiting as to the scope of invention which is to be given the full breadth of the claims appended and any and all equivalents thereof.

What is Claimed is:

1. A PGNA method for nondestructive assay of a non-homogeneous contaminant (50) in a container (36), said method comprising the steps of:
 - (a) employing an interrogation chamber (40) having a wall (42) made of a neutron-reflecting material;
 - (b) disposing a platform (30) within said interrogation chamber (40);
 - (c) employing at least one of:
 - (i) rotating (31) said platform (30), and
 - (ii) elevating (32) said platform (30);
 - (d) disposing said container (36) on said platform (30) and within said interrogation chamber (40);
 - (e) employing at least one neutron source (46) between the wall (42) of said interrogation chamber (40) and said container (36);
 - (f) irradiating said container (36) with a fast-neutron burst (44) from the neutron source (46) or as reflected by said wall (42), said fast-neutron burst characterized by a duty cycle and being thermalized and/or moderated by said container (36), with thermal neutrons (52) reacting with said container (36) and thereby effecting an emission of thermal-neutron induced gamma radiation (54) from said container (36);
 - (g) acquiring (10,11) gamma radiation data indicative of the number or intensity of gamma rays and energies of said gamma rays during a predetermined time interval, said acquiring step including a separate identification of thermal neutron-induced gamma rays, and wherein said gamma radiation data is acquired over only a fraction of said duty cycle; and

(h) analyzing (14,15,16) said gamma radiation data to detect the presence and determine the amount of said non-homogeneous contaminant (50) in said container (36).

2. The PGNAA method of Claim 1 further comprising:
rotating said platform on a rotating turntable (31).
3. The PGNAA method of Claim 2 further comprising:
rotating said rotating turntable (31) at a rotational speed of about 0 to 10 revolutions per minute.
4. The PGNAA method of Claim 1 further comprising:
elevating said platform (30) with an elevating platform (32), in order to elevate said container (36) and assay different sections thereof.
5. The PGNAA method of Claim 4 further comprising:
elevating said elevating platform (32) at an elevation speed of about 0 to 10 cm per minute.
6. The PGNAA method of Claim 1 further comprising:
employing both of said steps of rotating said platform (30), and elevating said platform (30).
7. The PGNAA method of Claim 6 further comprising:
rotating said platform (30) by about 360 degrees before elevating said platform (30).
8. The PGNAA method of Claim 6 further comprising:
rotating said platform (30) concurrently with elevating said platform (30).
9. The PGNAA method of Claim 1, wherein said analyzing step (14,15,16) includes interference-correction (15) to detect and/or correct for:
 - (a) signal interference from gamma rays emitted by other elements present in said container (36), and
 - (b) perturbation of interrogating neutron flux resulting from neutron-producing fissile materials and neutron poisons in said container (36).
10. The PGNAA method of Claim 1, wherein said contaminant (50) is a hazardous element.
11. The PGNAA method of Claim 1, wherein said container (36) is a sealed container (36).

12. The PGNA method of Claim 11, wherein said sealed container (36) is a 55 gallon waste drum (36).

13. The PGNA method of Claim 1, wherein said contaminant (50) is a plurality of RCRA metals; and said container (36) is a mixed-waste drum.

14. The PGNA method of Claim 1, wherein said analyzing step (14,15,16) includes computer modeling (16) to perform gamma spectral deconvolution.

15. The PGNA method of Claim 14, wherein said analyzing step (14,15,16) further includes a gamma spectral analysis program (16) to enhance system sensitivity for elemental detection.

16. The PGNA method of Claim 14, wherein said analyzing step (14,15,16) further includes adjoint transport calculations (16) to infer contaminant distribution through pattern recognition.

17. The PGNA method of Claim 1, further comprising:
employing as said neutron source (46) a plurality of neutron sources in order to produce significant thermal neutron fluxes at different average depths in said container (36); and
employing with said analyzing step (14,15,16) depth profiling analysis (14) of an element detectable by thermal neutron activation.

18. The PGNA method of Claim 1, further comprising:
said analyzing step (14,15,16) includes determining (14) whether said contaminant (50) is present within a prescribed depth range.

19. The PGNA method of Claim 18, further comprising:
computing a depth at which said neutrons have energy appropriate to initiate a reaction that produces gamma radiation indicative of said contaminant (50); and
generating depth profile data representing the amount of said contaminant (50) at a plurality of depth ranges in said container (36).

20. The PGNA method of Claim 1, wherein said contaminant (50) is a hazardous element.

21. The PGNA method of Claim 20, wherein said hazardous element is selected from the list consisting of mercury, cadmium, and lead.

22. A PGNA method for nondestructive assay of a non-homogeneous contaminant (74) or a homogeneous contaminant (28) in a container (68), said method comprising the steps of:

(a) employing an interrogation chamber (60,80) having an annular wall (62) made of a neutron-reflecting material (66), said wall (62) having a first portion and a second portion;

(b) disposing a neutron-moderating material (64) on the first portion of the wall (62) of said interrogation chamber (60);

(c) disposing a platform (30) within said interrogation chamber (60);

(d) employing at least one of:

(i) rotating (31) said platform (30), and

(ii) elevating (32) said platform (30);

(e) disposing said container (68) on said platform (30) and within said interrogation chamber (60);

(f) employing a plurality of neutron sources (70) between the wall (62,81) of said interrogation chamber (60) and said container (68);

(g) irradiating said container (68) with a fast-neutron burst from at least one of said neutron sources (70) or as reflected by the second portion of said wall (62), said fast-neutron burst characterized by a duty cycle and being thermalized and/or moderated by said container (68), with thermal neutrons reacting with said container (68) and thereby effecting an emission of thermal-neutron induced gamma radiation from said container (68);

(h) irradiating said container (68) with a thermal-neutron burst from at least one of said neutron sources (70) as moderated by said neutron-moderating material (64) on the first portion of the wall (62) of said interrogation chamber (60) and as reflected by said first portion of said wall (62), said thermal-neutron burst characterized by a duty cycle, with thermal neutrons reacting with a first layer (76) of said container (68) and thereby effecting an emission of thermal-neutron induced gamma radiation from said first layer (76);

(i) selectively acquiring (10,11):

(1) thermal-neutron induced gamma radiation from said container (68), or

(2) thermal-neutron induced gamma radiation from said first layer (76),

to provide gamma radiation data indicative of the number or intensity of gamma rays and energies of said gamma rays during a predetermined time interval, said selectively acquiring step including a separate identification of thermal neutron-induced gamma rays, and wherein said gamma radiation data is acquired over only a fraction of said duty cycle; and

(j) analyzing (14,15,16) said gamma radiation data to detect the presence and determine the amount of said non-homogeneous contaminant (74) or said homogeneous contaminant (28) in said container (68).

23. The PGNA method of Claim 22, further comprising:
employing as said neutron-reflecting material (66) a material selected from the list consisting of graphite, and bismuth.

24. The PGNA method of Claim 22, further comprising:
employing as said neutron-moderating material (64) a material selected from the list consisting of polyethylene, H₂O, D₂O, and graphite.

25. The PGNA method of Claim 22, further comprising:
disposing a neutron-reflecting material (86) on the second portion of said wall (81) opposite said first portion of said wall (81).

26. The PGNA method of Claim 22, further comprising:
disposing no material on the second portion of said wall (62) opposite said first portion of said wall (62).

27. A PGNA apparatus (1) for nondestructive assay of a non-homogeneous contaminant (50) in a container (36), said apparatus (1) comprising:
an interrogation chamber (40) having a wall (42) made of a neutron-reflecting material;

a platform (30) for supporting said container (36) within said interrogation chamber (40);

means (17) for rotating and/or elevating said platform (30);

at least one neutron source (46) between the wall (42) of said interrogation chamber (40) and said container (36);

means (7,8) for irradiating said container (36) with a fast-neutron burst from said neutron source (46) or as reflected by said wall (42), said fast-neutron

burst characterized by a duty cycle and being thermalized and/or moderated by said container (36), with thermal neutrons reacting with said container (36) and thereby effecting an emission of thermal-neutron induced gamma radiation (54) from said container (36);

at least one gamma ray detector (56,92);

means (10,11) for acquiring gamma radiation data from said gamma ray detector (56) indicative of the number or intensity of gamma rays and energies of said gamma rays during a predetermined time interval, including a separate identification of thermal neutron-induced gamma rays, and wherein said gamma radiation data is acquired over only a fraction of said duty cycle;

means (14,15,16) for analyzing said gamma radiation data to detect the presence and determine the amount of said non-homogeneous contaminant (50) in said container (36).

28. The PGNAA apparatus (1) of Claim 27, wherein said gamma detector (92) includes a shield (97).

29. The PGNAA apparatus (1) of Claim 28, wherein the shield (97) of said gamma detector (92) is a ^6Li -bearing shield (97).

30. The PGNAA apparatus (1) of Claim 29, wherein said ^6Li -bearing shield (97) is made of lithium-6 fluoride.

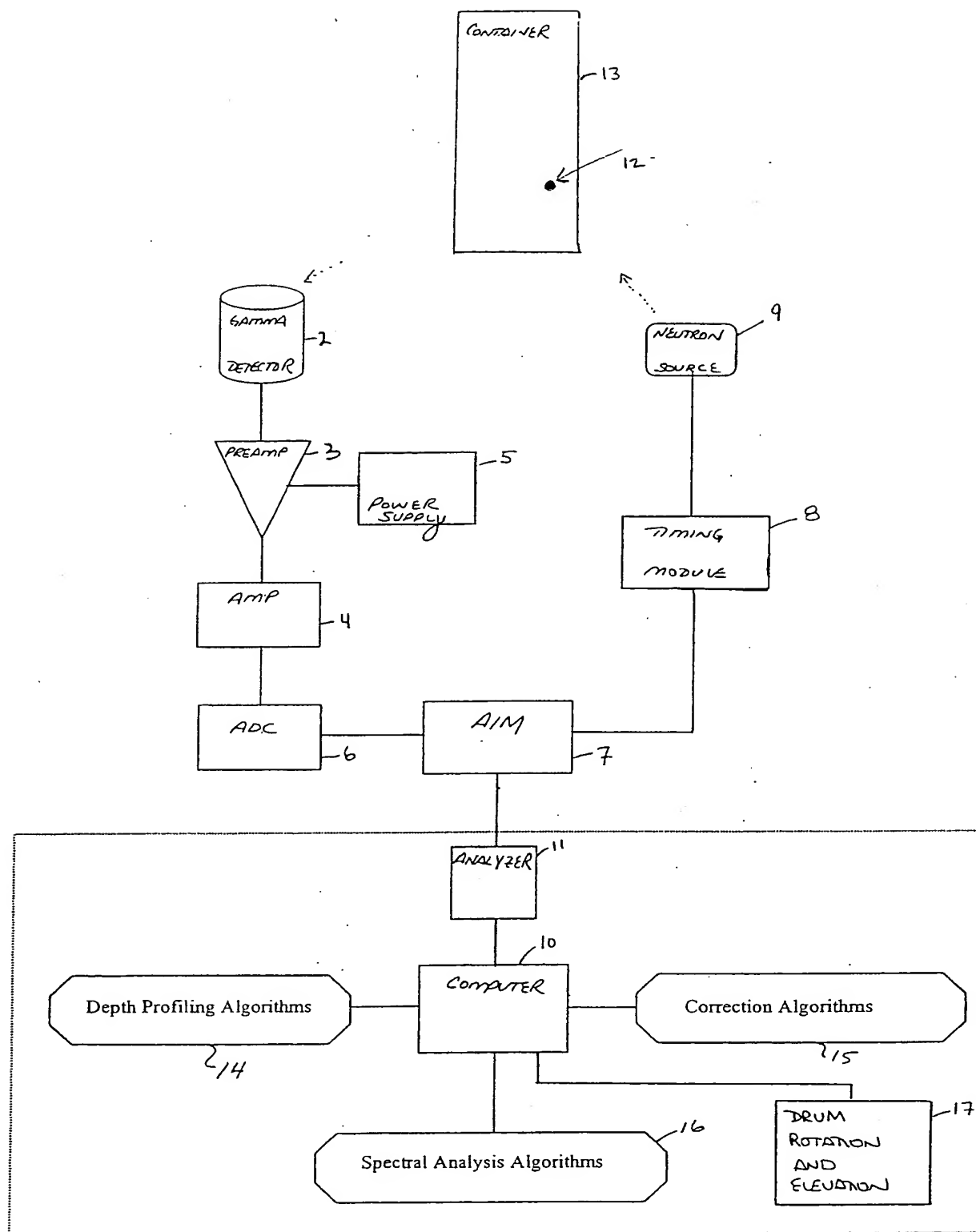
31. The PGNAA apparatus (1) of Claim 29, wherein said ^6Li -bearing shield (97) is disposed around a detector (94); and wherein a moderating and/or shielding material (98) is disposed around said ^6Li -bearing shield (97).

32. The PGNAA apparatus (1) of Claim 27, wherein said moderating and/or shielding material (98) is selected from the list consisting of bismuth, and tungsten.

33. The PGNAA apparatus (1) of Claim 27, wherein said at least one neutron source (46) is a plurality of neutron sources (46).

34. The PGNAA apparatus (1) of Claim 27, wherein said at least one gamma ray detector (56) is a plurality of gamma ray detectors (56).

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FIG. 1

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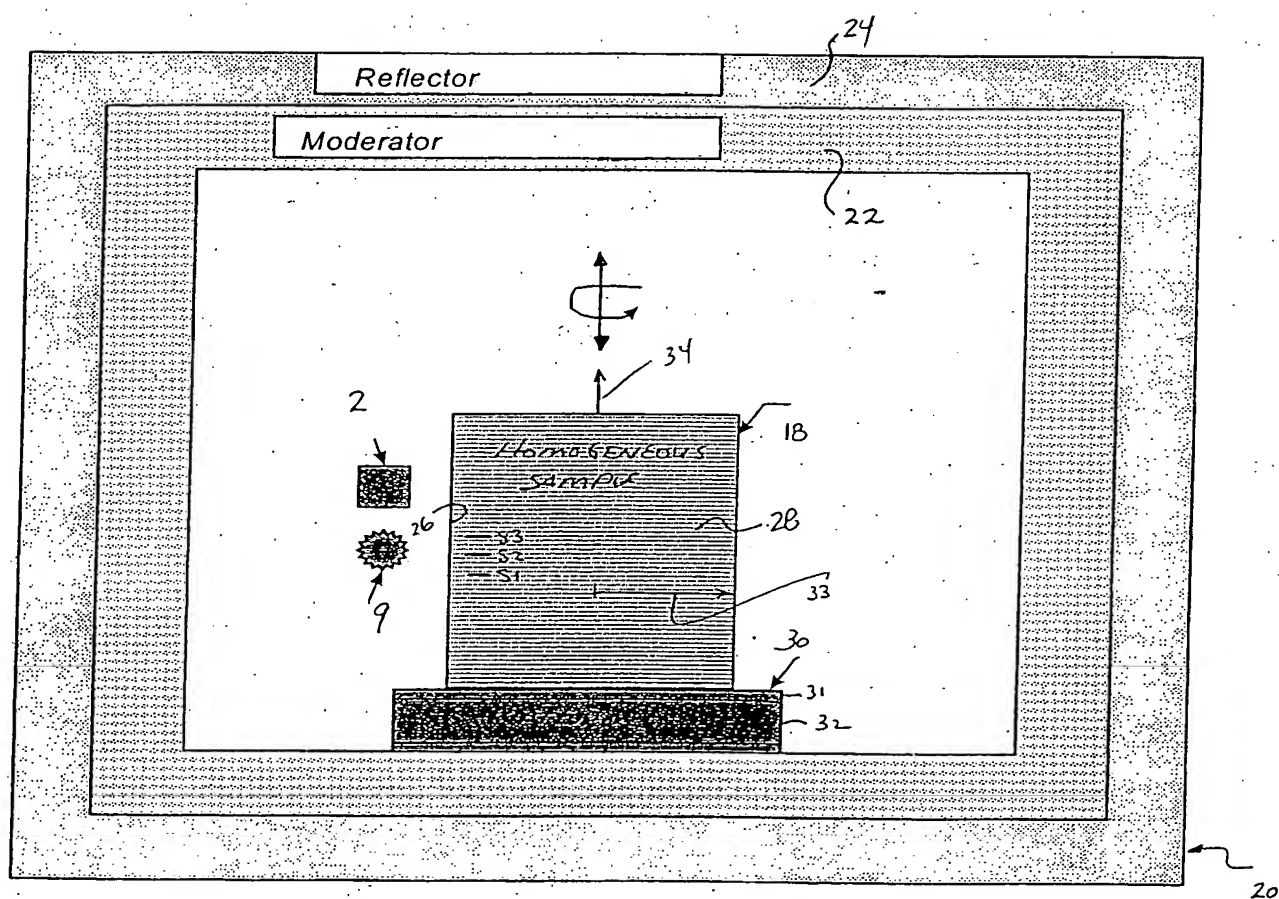


FIG. 2

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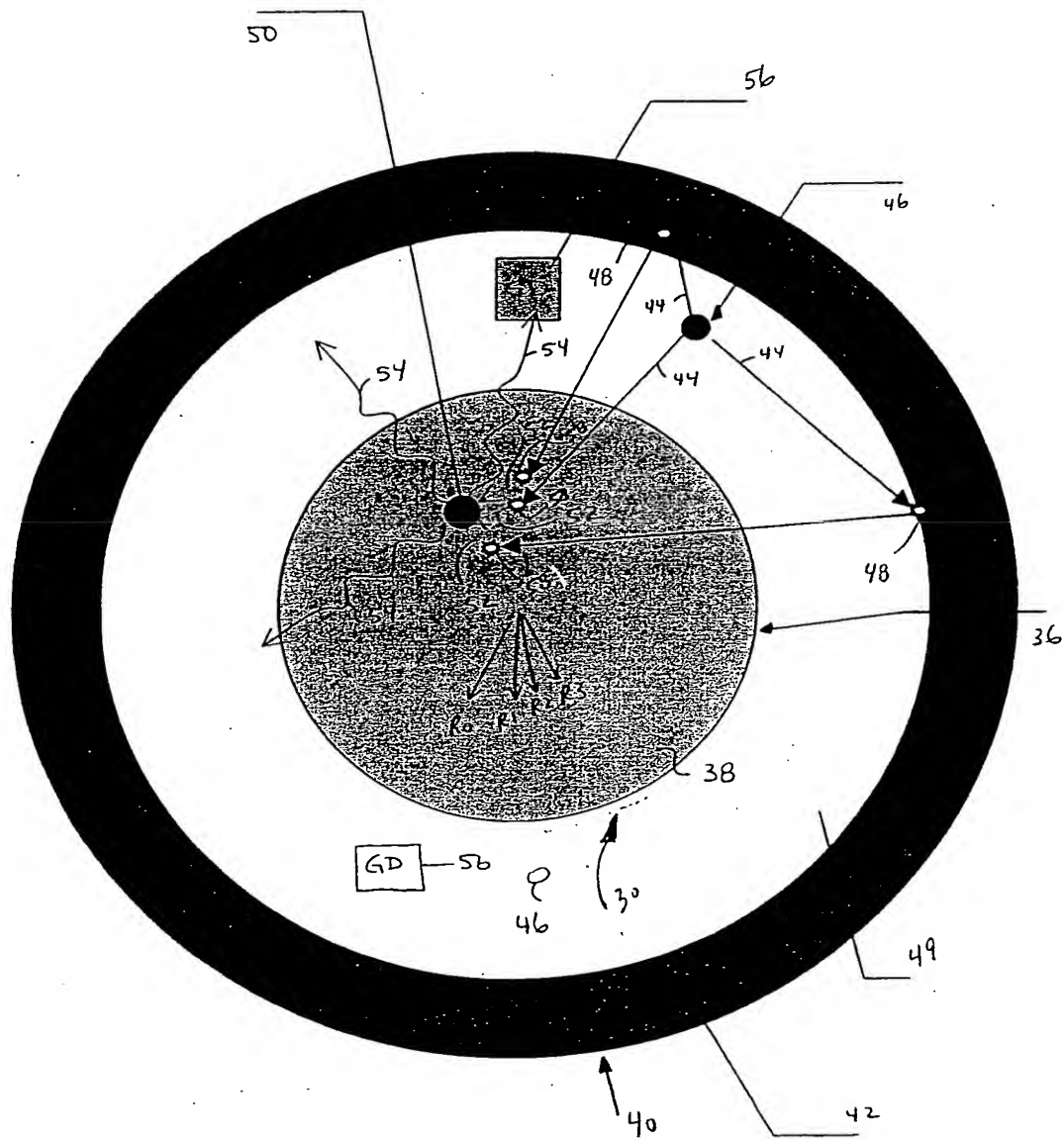


FIG. 3

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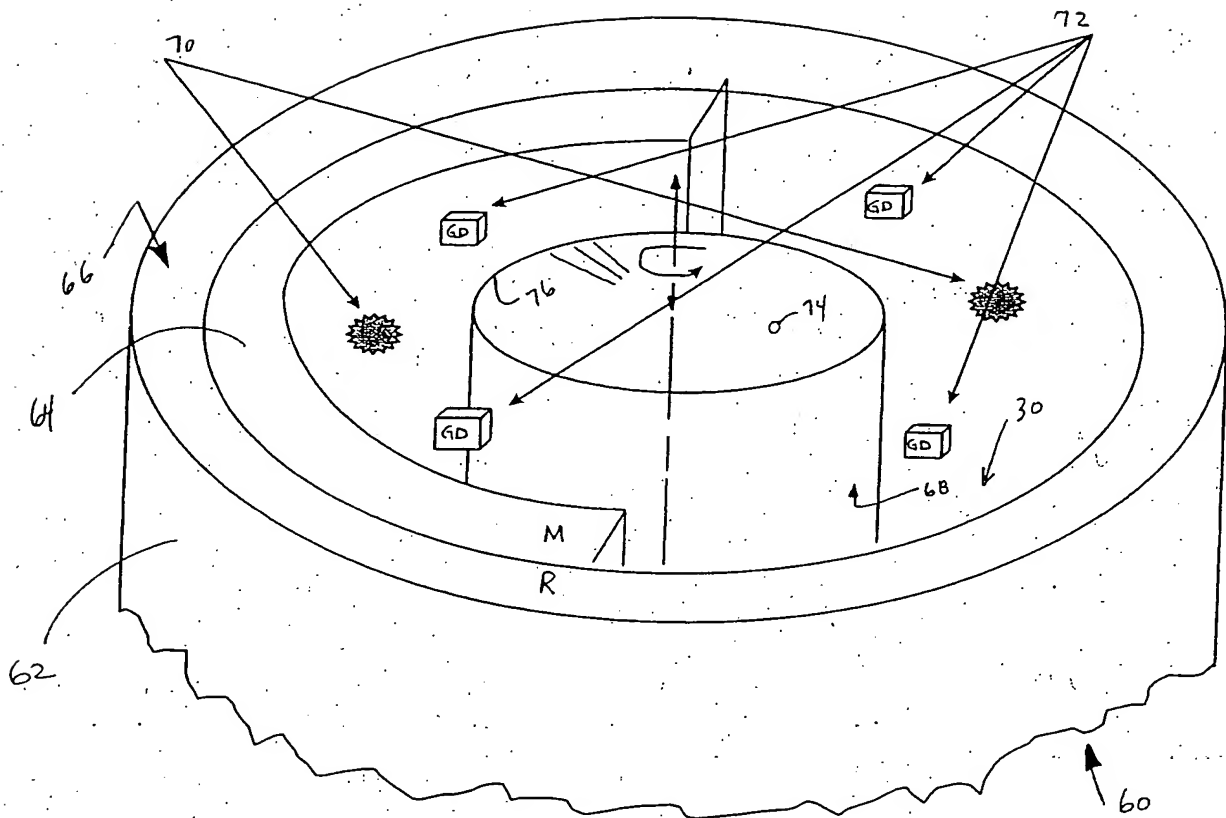


FIG. 4

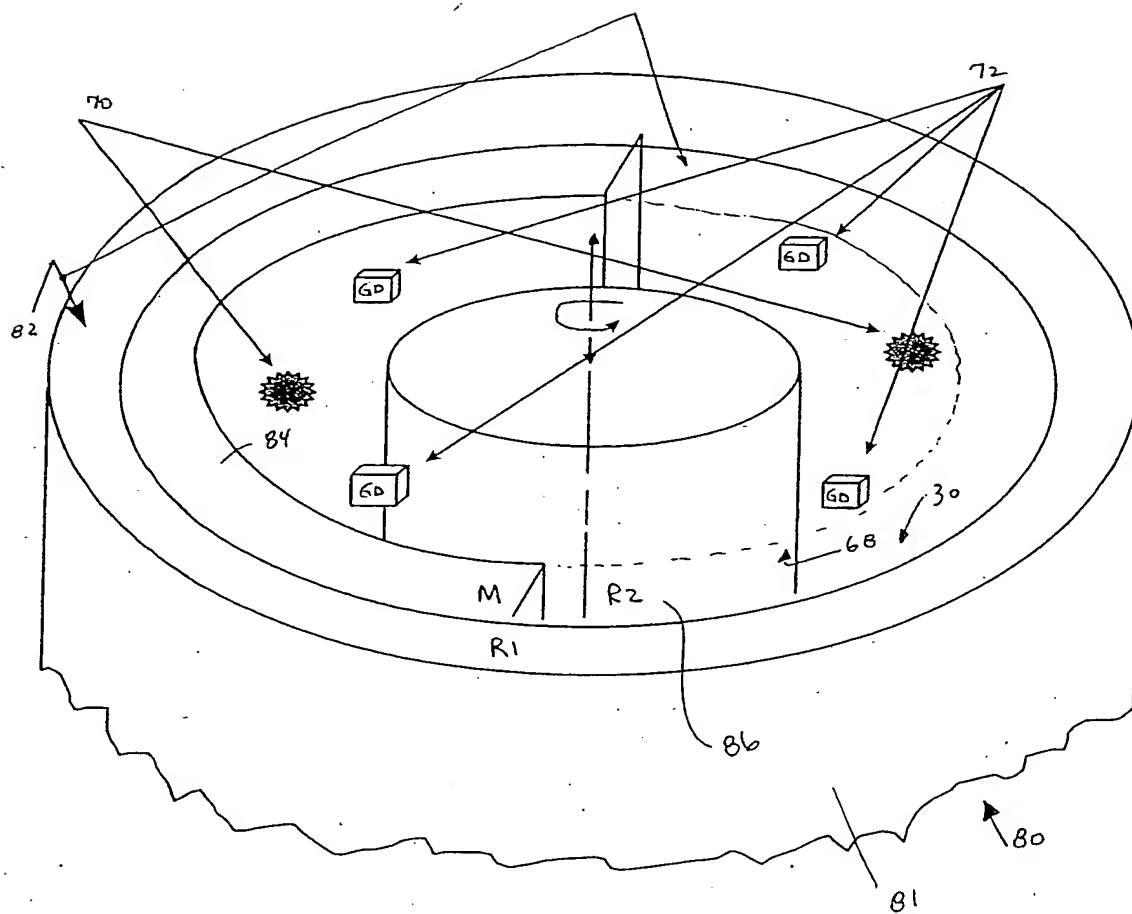


FIG. 5

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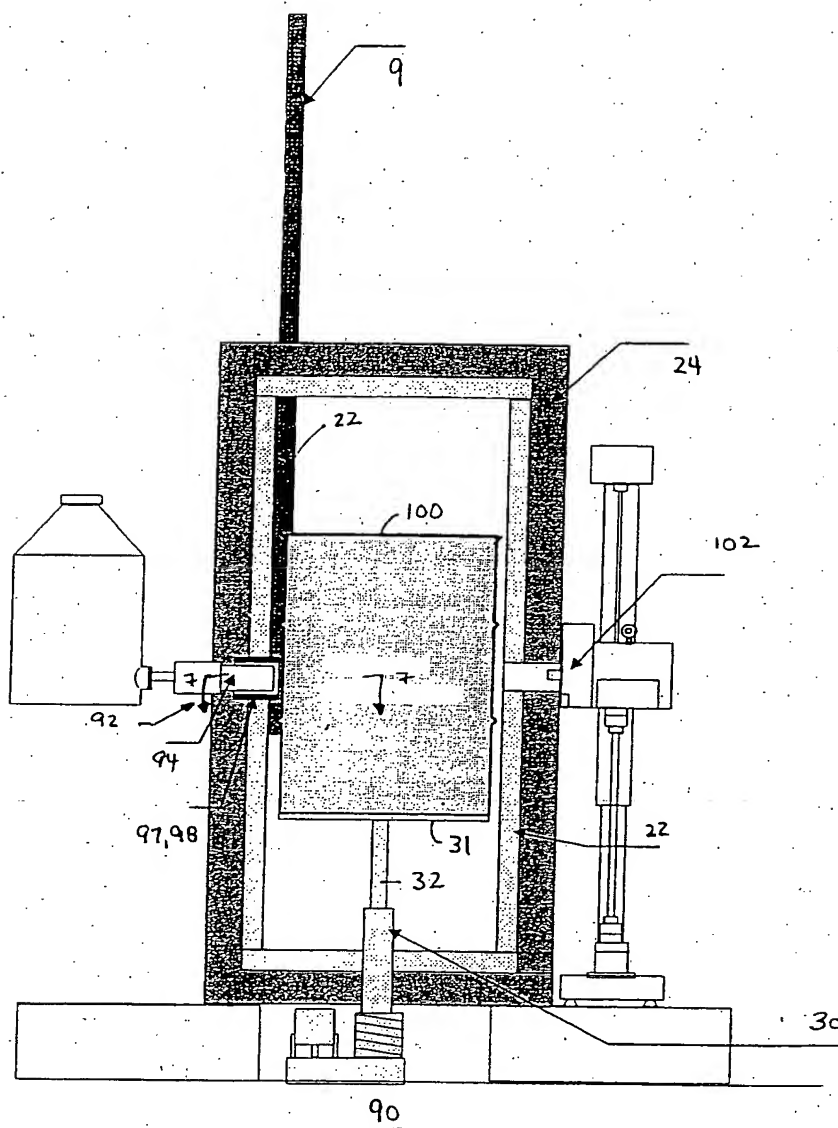


FIG. 6

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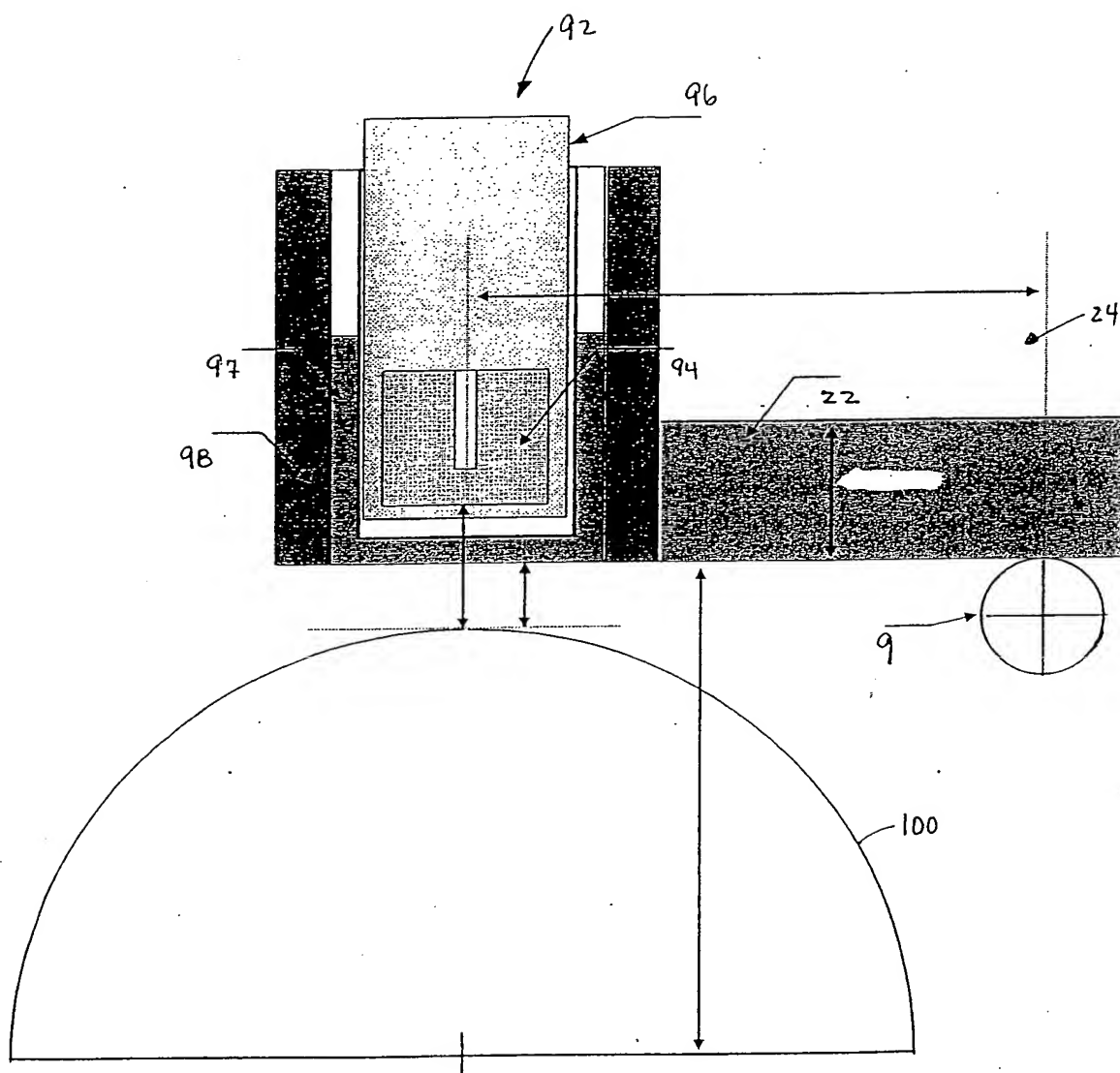


FIG. 7

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